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Polyoxymethylene/Thermoplastic Polyurethane Blends Compatibilized with Multifunctional Chain Extender

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ABSTRACT: Novel compatibilized polyoxymethylene/thermoplastic polyurethane (POM/TPU) blends are successfully developed using multifunctional chain extender, Joncryl ADR-4368, as the compatibilizer. The outstanding compatibilization efficiency of Joncryl on POM/TPU blend was demonstrated by its even higher mechanical properties with only 0.5 wt % of Joncryl than those with 5 wt % of three commonly used compatibilizers. Addition of only 0.5 wt % Joncryl can double the impact strength and significantly improve its tensile strength and flexural strength for POM/TPU (75/25) blend. SEM images show that Joncryl can reduce TPU particle size and enhance the interfacial interactions between POM and TPU. The interparticle distance of TPU in POM/TPU/Joncryl blends was calculated as 0.2 μm, quite close to the critical matrix ligament thickness of POM/TPU blends (0.18 μm). The impact force profile vividly shows that the addition of Joncyl in POM/TPU blends can dramatically increase the total impact energy absorbed by this blend system and enhance the interfacial interactions between POM and TPU. © 2012 Wiley Periodicals, Inc. J. Appl. Polym. Sci. 000: 000–000, 2012

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INTRODUCTION

Polyoxymethylene (POM) is one of the widely used engineering plastics with a unique balance of mechanical, thermal, chemical, and electrical properties. Structurally, POM has a typical helical chain structure composed of alternating sequence of gauche C—O bonds. This regular conformation endows POM with high crystallinity (ca. 60–80%) to form large spherical crystals during melting process. All these properties along with its good processability offer POM wide applications in machinery, automobiles, and electric/electronic industries.[1–3] However, the brittleness at room temperature and low temperatures accompanied with its high crystallinity often exert great limitation to POM for certain applications.

To improve the impact toughness of POM and extend its application range, considerable efforts have been made on the toughening of POM.[4–8] Among the elastomers used, thermoplastic polyurethane (TPU) is so far the best toughening agent and can simultaneously enhance both the elongation and thermal stability of POM due to its good compatibility with POM,[8–14] which is attributed to the possible formation of hydrogen bond between part of POM ether bonds and TPU.[10] The impact strength of POM/TPU blends can be significantly improved with higher addition of TPU (content over 30 wt %), and the toughening mechanism is explained as the formation of a co-continuous morphology at high TPU addition.[11] In order to further improve the compatibility of POM/TPU, effective compatibilizers were added to enhance the interfacial interactions between POM and TPU. Examples include diphenylmethane diisocyanate (MDI),[8] polystyrene-*block*-poly(ethylene- butylene)-*block*-polystyrene grafted with maleic anhydride (SEBS-*g*-MAH),[9] and ethylene-propylene-diene grafted with maleic anhydride (EPDM-*g*-MAH).[15] The use of appropriate compatibilizer is beneficial for the reduction of TPU particle size, improved dispersion of TPU in POM, formation of good interface to enhance the interface bonding, and therefore increase of impact strength of the resulted compatibilized blends.

For elastomer-toughened rigid plastic blends, various toughening mechanism have been proposed, including crazing, cavitation, and shear yielding.[9] Among them, the critical matrix ligament thickness (L_c) theory has found its applicability as the shear yielding mechanism account for nylon/EPDM,[16] PP/

EPDM,[17] and POM/TPU toughened polymer blends.[9] In this model, the impact toughness correlated with the phase morphology, i.e., L_c is the only parameter determining the brittle–ductile (B–D) transition. Only when the matrix ligament thickness (L) is smaller than L_c can shear yielding of matrix ligament exist and a B–D transition of the blends occurs. For given blends, L_c is independent of the weight fraction and particle size of elastomer. In the case of POM/TPU blends, no "supertough" behavior has been observed due to its very small L_c value (0.18 μm).[18] To achieve toughened POM/TPU, efforts need to be made to reduce TPU interparticle distance close to L_c of POM.

Recently, researchers have explored multifunctional chain extender, Joncryl ADR-4368, to effectively rebuild the molecular weight and physical properties of degraded thermoplastics such as PET and PBT.[19–21] Mehrabzadeh et al.[8] successfully developed toughened POM/TPU blends with bifunctional chain extender (MDI). So far, there is no study on toughening POM/TPU blending via multifunctional chain extender. The investigations of POM/TPU system have mainly focused on the influence of TPU content on mechanical properties, the selection of compatibilizer, interfacial reaction, as well as rheological properties.[9–15,22]

This article reports our endeavor in developing toughened POM/TPU blends with the chain extender, Joncryl ADR-4368. Its efficacy in toughening POM/TPU blends was evaluated in reference to MDI, EPDM-*g*-MAH and poly(ethylene-octene) grafted with maleic anhydride (POE-*g*-MAH).[23] The mechanical properties, morphology, interparticle distance, and impact force profile of different compatibilized POM/TPU blends were compared with better understand the compatibilization efficacy of Joncryl.

EXPERIMENTAL

Materials

Polyoxymethylene (POM, Lucel N109-LDS) was obtained from LG Chemical, Korea, has a melt flow index (MFI) of 13 g/10 min (190°C under 2.16 kg load). The thermoplastic polyurethane (TPU, Skytane-S180A) used in this study was obtained from Sunkyong Industries, Korea. Diphenylmethane diisocyanate (MDI) was obtained from Merck. EPDM-*g*-MAH (CG700), obtained from Chen-Guang Chemical Institution, has a grafting ratio of 0.92 wt % and MFI of 5 g/10 min (230°C under 2.16 kg load). POE-*g*-MAH (DFDA 1373) was purchased from UCC, USA. Chain extender-Joncryl ADR-4368, procured from BASF, has a M_w of 6800, an epoxy equivalent weight of 285 g/mol, a functionality (f_n) greater than 4 and tailored polydispersity (PDI>3). Joncryl ADR-4368 was referred as Joncryl in this study unless specified. For all blends, POM/TPU composition was fixed as 75/25, where 0.5 wt % weight content of Joncryl was used, while 5 wt % for commonly used compatibilizer (including MDI, EPDM-*g*-MAH, and POE-*g*-MAH).

Preparation of Compatibilized POM/TPU Blends

All blends were melt-mixed in a corotating twin-screw extruder (SLF-35B, L/D = 30, Keqiang Polymer Engineering Company, China). In the extrusion step, the barrel temperature profile was set as 160, 170, 185, 185, and 180°C from hopper to die and a screw speed of 200 rpm was used. The extrudates were immediately quenched in water and subsequently cut into pellets. The standard regular bars and dumbbell-shaped specimens for flexural, impact and tensile properties testing were injection-molded using a JPH-120 injection-molding machine. The temperature profile for injection molding was set as 175, 185, 190, and 190°C from hopper to die.

Mechanical Properties

The notched Izod impact strengths were determined with a XJJ-5 pendulum impact tester at 25°C according to ASTM D256. The impact force profile was also recorded on this impact tester. The average value of six to eight measurements was reported for each blend composition. The tensile strength and flexural strength were measured as per ASTM D638 and ASTM-D790 methods, respectively. The measurements were carried out on an Instron-3211 universal tensile tester. The crosshead speed was set as 10 mm/min for tensile tests and 2 mm/min for flexural measurements. The values of both mechanical parameters were calculated as average over six to eight specimens for each composition.

Scanning Electron Microscopy

The morphological characteristics were examined using scanning electron microscopy (SEM). Prior to examining, the samples were first fractured along the direction perpendicular to the melt flow direction in liquid nitrogen. The fracture surface was then coated with a thin layer of gold. The fracture morphology was observed with a JEOL JSM-6360LV scanning electron microscopy, using an acceleration voltage of 20 kV.

SEM images obtained were subsequently segmented and subjected to digital analysis using ImageTool 3.0 software to elucidate the statical size distribution of dispersed compatibilizer droplets.

RESULTS AND DISCUSSION

Mechanical Properties of POM/TPU Compatibilized Blends

The toughening efficacy of novel chain extender-Joncryl for POM/TPU (75/25) blends is compared with commonly used compatibilizers MDI, EPDM-*g*-MAH and POE-*g*-MAH. The compatibilization of Joncryl for POM/TPU blends can be realized by its ability to improve the interfacial interaction between of POM and TPU via reaction blending. The mechanisms of epoxy based Joncryl in reactive extrusion have been discussed at length in literature.[19,20] In polyamides like TPU, amidation of terminal amine groups is by far the dominant mechanism. For POM, etherification of backbones' end hydroxy groups with epoxy group of Jocryl governs the chain extension process (Figure 1). Clearly, in each one of these systems extensive long chain branching can result in undesirable crosslinking when the extender has a f_n greater than two.[19] Hence, if Joncryl was employed to enhance the interfacial interaction for POM/TPU blends its content must be controlled to reach given extent of reaction.[19]

The impact strength of various POM/TPU/compatibilizer blends is compared in Figure 2. The Izod impact strength of POM/TPU blends was found to increase dramatically with the addition of compatibilizer. The addition of 0.5 wt % Joncryl brings about over 100% increment in impact strength for POM/TPU

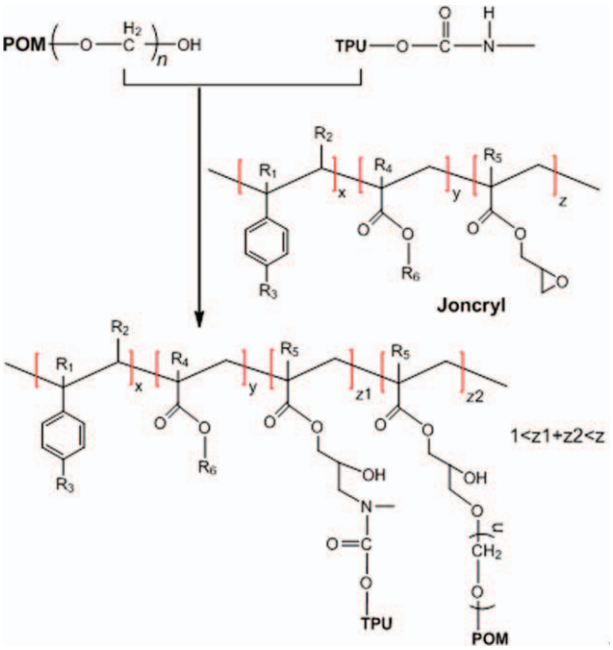

Figure 1. Compatibilization mechanism of multi-functional extenders Joncryl for POM/TPU blends, leading to long chain branching. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

blends, where an Izod impact strength of 32.1 ± 0.5 kJ/m^2 was obtained, compared with 16 ± 0.2 kJ/m^2 for pure POM/TPU (75/25) blends.[9] The addition of 5 wt % generally used compatibilizers including MDI, EPDM-*g*-MAH, and POE-*g*-MAH, also result in 70–90% enhancement in the impact strength of POM/TPU blends.

Among them, Joncryl presents as the most effective compatibilizer and MDI as the second. This behavior can be explained by their chemical structures and compatibilization mechanisms. EPDM-*g*-MAH and POE-*g*-MAH are both mono-functional compatibilizers, which can react with hydroxyl groups of POM using their contained maleic anhydride group to improve the interfacial compatibility between POM and TPU. MDI is a difunctional chain extender with two isocyanate ending group, which can react not only with the end hydroxyl groups of POM chains but the urethane groups in the TPU chains. This coupling leads to increased interfacial adhesion between POM and TPU, and thus better compatibility.[8] Joncryl is a multiepoxide styrene-acrylic polymer chain extender, whose epoxy functional groups can undergo ring-opening reaction with hydroxyl groups of POM chains and urethane groups of TPU chains.[19–22] Since Joncryl is multifunctional ($F_n > 4$) chain extender, 0.5 wt % addition is enough to increase the interface adhesion between POM and TPU. The impact strength can be significantly enhanced for POM/TPU/Joncryl compatibilized blends. It should be noted that increasing the addition of Joncryl in POM/TPU(75/25) system to ca. 5 wt % could result in decreased impact strength though enhanced tensile and flexural strengths in comparison to the current studied 0.5 wt % Joncryl compatibilized blends.

The effect of Joncryl on the tensile strength and flexural strength (stiffness) is shown in Figure 3 and compared with other three compatibilizers. Similar behavior was observed for all four blend systems. However, Joncryl and MDI compatibilized POM/TPU blends presented better stiffness than EPDM-*g*-MAH and POE-*g*-MAH, mainly due to their improved compatibilization of POM and TPU by using bi- or multifunctional chain extender.

A close look at the tensile strength and flexural strength (stiffness) of Joncryl and MDI compatibilized POM/TPU blends, one can find that these two blends presented the highest stiffness (both tensile and flexural strength). More importantly, 0.5 wt % Joncryl exhibited almost equal compatibilization efficacy as 5 wt % MDI does. For EPDM-*g*-MAH and POE-*g*-MAH, though both mono-functionalized with maleic anhydride grafting, POE-*g*-MAH compatibilized POM/TPU blends showed slightly better flexural and impact strength than EPDM-MAH, probably due to better miscibility of POE polymer chain with POM.

Fracture Morphology and Matrix Ligament Thickness

To better understand the compatibilization efficacy of Joncryl, SEM images of notched impact fracture samples for different compatibilized POM/TPU blends were shown in Figure 4. Relatively smooth surfaces were observed for POM/TPU blends, not matter what compatibilizer was used. The typical "island-sea" morphology is observed for the toughened POM/TPU blends, a similar morphology observed in ethylene-styrene interpolymer compatibilized polystrene/polyethylene[24,25] and styrene-*b*-ethylene/butylene-*b*-styrene triblock copolymer compatibilized polyphenylene sulfide/nylon 66 blend systems.[26] Thus, crazing or shear yielding cannot be considered as the toughening mechanism for POM/TPU/compatibilizer blends.

As observed, the dispersed TPU particles present different domain size and distribution with the addition of different compatibilizers. The TPU particle size and size distribution directly obtained from the SEM images were subject to digital analysis,

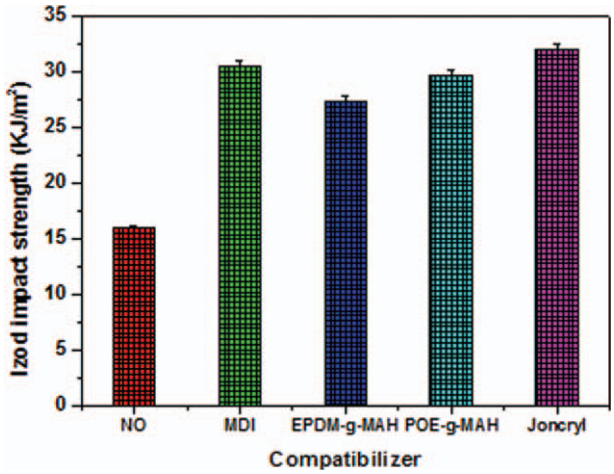

Figure 2. The Effect of compatibilizers on impact strength of POM/TPU (75/25) blends. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

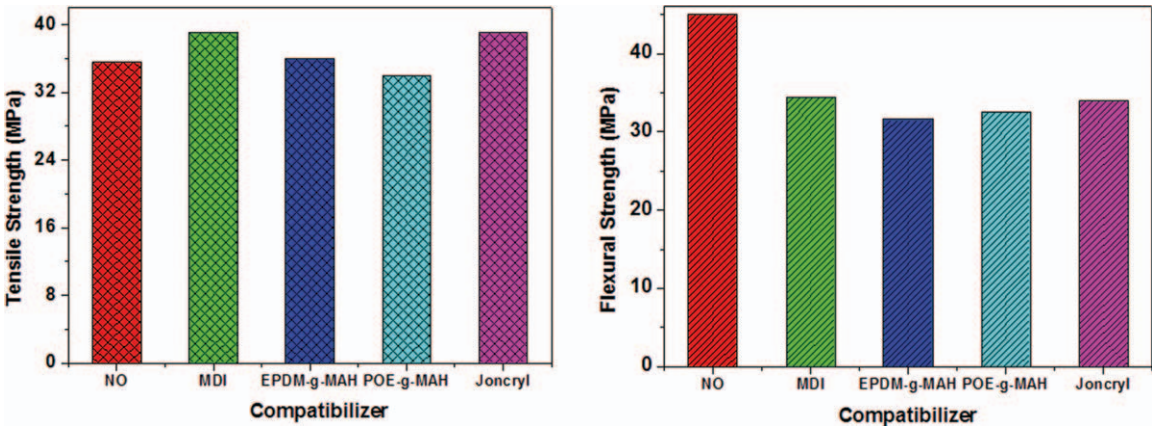

Figure 3. The effect of compatibilizers on tensile strength (a) and flexural strength (b) of POM/TPU (75/25 w/w) blends. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

with resulting histogram shown in Figure 5. The histograms of statistical size distribution of TPU droplets were analyzed with reversible aggregation model.[27,28] According to the model, the stationary distribution $h(s)$ of the planar size s of the microstructural entities can be expressed as follows

$$h(s) = \sum_{i=1}^{N} a_i s_i^2 \exp\left(-\frac{s_i \Delta u_{0i}}{kT}\right) \quad (1)$$

where a is normalizing factor, $\triangle u_{0i}$ is the aggregation energy, k is the Boltzmann constant, T is the absolute temperature

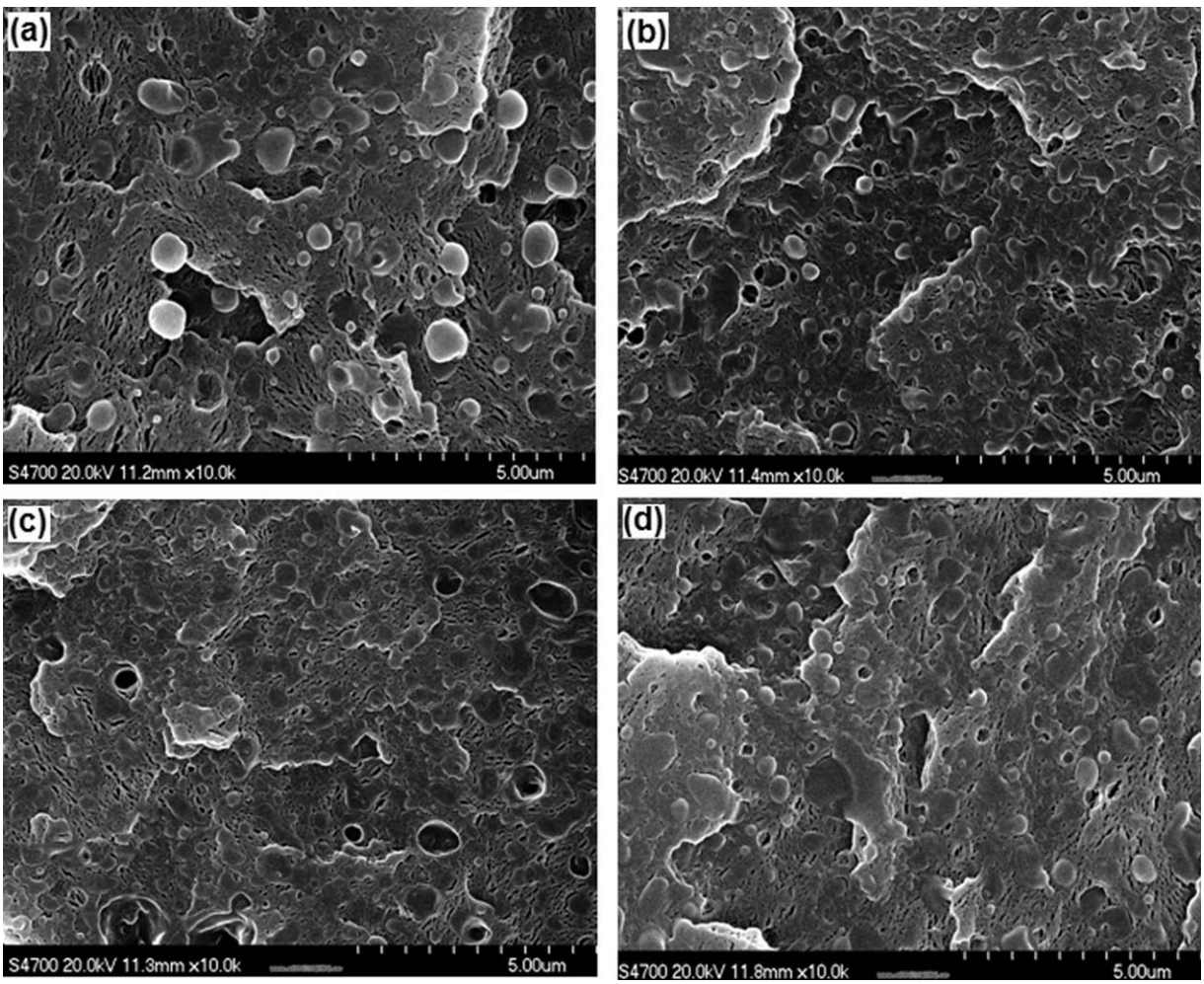

Figure 4. SEM micrographs showing cryogenically fractured surface features of POM/TPU blends compatibilized with (a) MDI, (b) EPDM-*g*-MAH, (c) POE-*g*-MAH, and (d) Joncryl. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

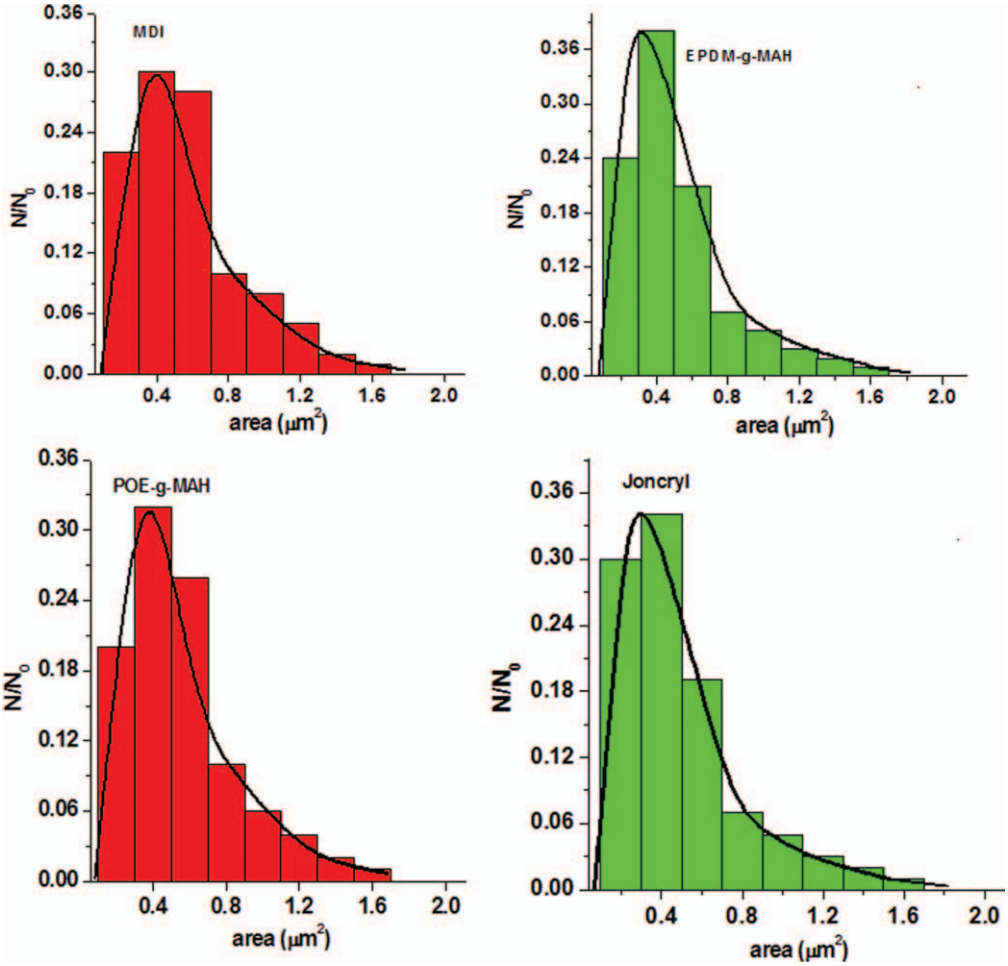

Figure 5. Statistical data of TPU particle size from SEM micrographs of Figure 4. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

and kT is the thermal fluctuation. N stands for the total number of statistical ensembles of the entities. Equation (1) allows the determination of mean entity area $<s_i>$ and linear mean size (diameter$<d_i>$) of TPU droplets by following eq. (2)

$$< d_i >= 2\sqrt{\frac{< s_i >}{\pi}} \quad (2)$$

The fitting curves with eq. (1) are shown in Figure 5. The successful analytical description indicates that the TPU droplets formed two super-imposed thermodynamically optimized statistical ensembles of primary and coalescent droplets all across the phase separation in the studied POM/TPU blends.

The average size of the droplets can be obtained with eq. (2) and summarized in Table I. Whatever compatibilizer was used, TPU dispersed phase exhibited small particle size, with an average size ranging from 0.28 to 0.42 μm, which is much smaller than that in pristine POM/TPU blends (average TPU particle size $\sim$ 1 μm).[9] In POM/TPU/Joncryl blends, TPU particles presented smallest dimension (average diameter 0.28 μm) and narrowest size distribution, while largest TPU particles (average diameter 0.42 μm) were observed in POM/TPU/MDI blends. The much decreased particle size of TPU in these compatibilized POM/TPU blends indicates that greater interaction between POM and TPU were achieved with the addition of compatibilizer. Among them, Joncryl demonstrated as most efficient compatibilizer in enhancing the interfacial interaction for POM/TPU blends.

According to Wu's proposed critical matrix ligament theory, the critical matrix ligament thickness (L_c) is the only parameter dominating the brittle-ductile transition.[16] Kawaguchi et al.[9] explored the interfacial reaction and its influence on phase morphology and impact properties of end-group modified-POM/TPU blends. The results indicated that the impact strength depends not only on the interparticle distance but also on the interfacial interactions between POM and TPU. In our study, chain extenders Joncryl and MDI demonstrated their greater capability in enhancing the interfacial interactions between POM and TPU than POE-*g*-MAH and EPDM-*g*-MAH. To reveal the effect of interparticle distance of TPU particles on the impact strength of POM/TPU/compatibilized blends, the matrix ligament thickness (L) is calculated as follows:[16]

Table I. Statistical Data of TPU Particles in POM/TPU/Compatibilizer Blends

Compatibilizer	MDI	EPDM-*g*-MAH	POE-*g*-MAH	Joncryl
Sum of dispersed particles	119	209	188	159
Sum of particles (diameter≤0.5 μm)	96	191	167	142
Sum of particles (diameter ≥1 μm)	23	18	21	17
Average diameter, *d* (μm)	0.42	0.32	0.38	0.28
W_f	0.238	0.238	0.238	0.249
V_f	0.267	0.267	0.267	0.279
L (μm)	0.31	0.24	0.28	0.21

$$L = d\left[\left(\pi/6V_f\right)^{1/3} - 1\right] \quad (3)$$

$$V_f = \frac{\rho_m W_f}{(\rho_m - \rho_c)W_f + \rho_c} \quad (4)$$

where d is the average TPU particle size, V_f the particle volume fraction of TPU, ρ_m the density of POM (1.4 g cm^{-3}), W_f the weight ratio of TPU (here is 0.238 = 25/105), and ρ_c the density of TPU (1.2 g cm^{-3}). TPU particles are assumed to disperse in POM matrix in random distribution.

The matrix ligament thickness (L) or interparticle distance for different POM/TPU/compatibilizer blends can be calculated from eq. (3) by using TPU particle size obtained directly from SEM images, and the results are summarized in Table I. The L values for POM/TPU/MDI, POM/TPU/EPDM-*g*-MAH and POM/TPU/POE-*g*-MAH (weight ratio 75/25/5) blends are 0.31, 0.24, and 0.28 μm, respectively. Totally, 0.5 wt % Joncryl compatibilized POM/TPU blend presented the lowest L value as 0.2 μm, which is quite close to the reported L_c value of 0.18 μm.[18] Our results indicate that the impact strength of POM/TPU/compatibilizer blends depends on not only the interparticle distance of dispersed TPU phase but also the interfacial interactions between POM and TPU. And both TPU interparticle distance and interfacial interactions between POM and TPU are strongly determined by the structure of compatibilizer used. In our cases, Joncryl demonstrated as the most effective compatibilizer for POM/TPU blends when considering its low addition and achievement of both improved toughness and stiffness for the resulted blends.

Impact Force Profile

To better understand the effect of TPU particle size and interfacial interactions on the impact strength of different POM/TPU/compatibilizer blends, the impact force profile was recorded and compared to get the insight on the outstanding compatibilization efficacy of Joncryl. In the typical pendulum impact test, characteristics of impact force profile[29] are defined as Figure 6. The impact force increases after the pendulum hits the notched specimen while decreases after fracture initiates within the specimen. The impact force profile oscillates subsequently as a result of fixture vibrations, which is determined by the natural frequency of impact test system and the rigidity of the pendulum. The first half-sine part of the impact force profile is generally considered, which represents the structural behavior of the notched specimens from the initiation of impact load till fracture completes. Characteristics for impact force profile include the maximum impact force (F_{max}), the duration of the first half-sine part of the impact force profile (T) and total impact energy absorbed. Total impact energy absorbed is defined as the area below the first half-sine part of the impact profile, which represents the toughness of the sample.

In our investigation, the impact force profiles were recorded for four POM/TPU/compatibilizer blends, as shown in Figure 7. Among the four compatibilized blends, POM/TPU/Joncryl [Figure 7(d)] and POM/TPU/MDI [Figure 7(a)] blends present higher F_{max} (ca. 235 N), indicating higher interfacial interaction existing in the polymer blends. POM/TPU/Joncryl blends also showed obviously longer impact force profile than the other three systems, with impact duration of 1.36 ms and even an extra deformation time of 0.64 ms observed in Figure 7(d). The rest three compatibilized blends displayed quite similar impact duration (1.2–1.3 ms). As a result, Joncryl compatibilized POM/TPU blend absorbed the highest impact energy due to its longest impact duration and highest impact force. Therefore, this blend exhibited the best impact strength. POM/TPU/MDI blends displayed higher impact strength than maleic anhydride grafted EPDM or POE compabilized POM/TPU blends.

CONCLUSIONS

Oligomer chain extender Joncryl is found to be the most effective compatibilizer for POM/TPU blends in comparison with the commonly used copolymer compatibilizers including MDI, EPDM-*g*-MAH, and POE-*g*-MAH. With the addition of 0.5 wt % Joncryl, POM/TPU/Joncryl blends harvested both higher toughness and stiffness than other three counterpart blends with 5 wt % compatibilizers. The impact strength of POM/TPU blends depended on not only the interparticle distance but also

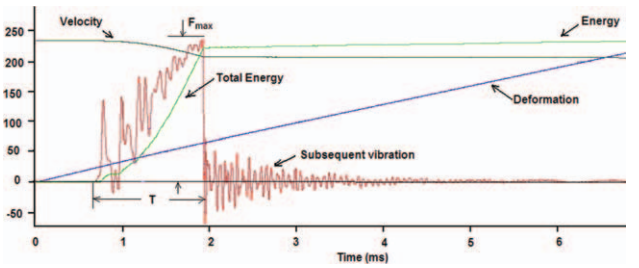

Figure 6. Characteristics of impact force profile. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

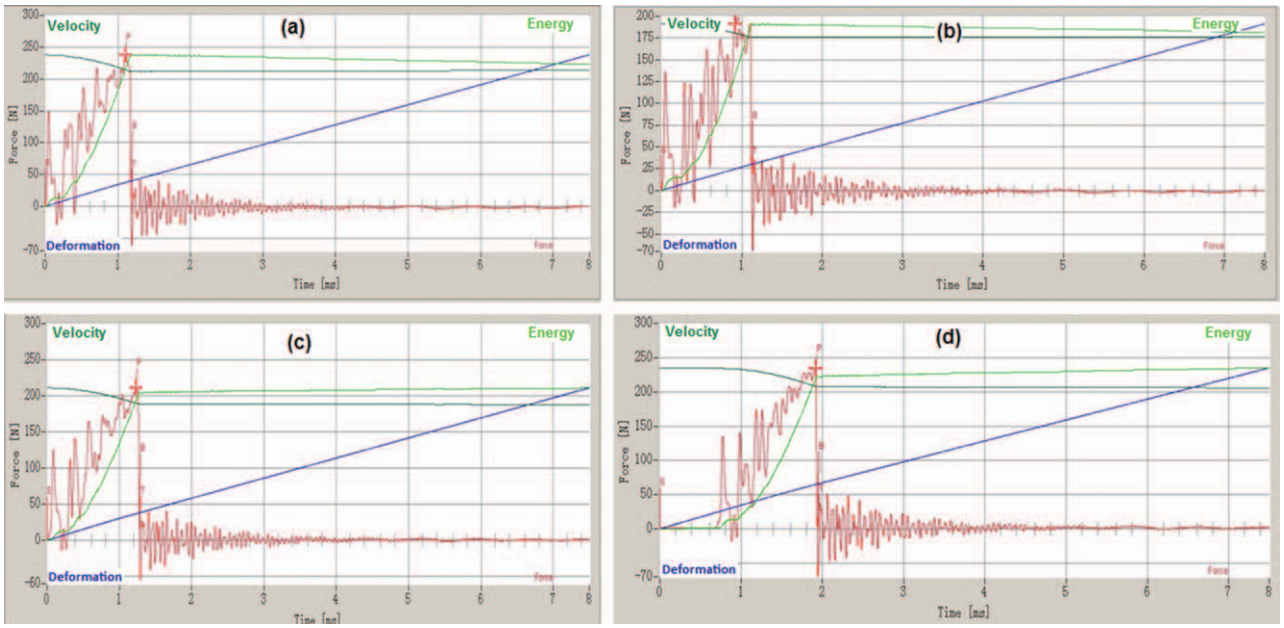

Figure 7. Energy-force profile during impact process for POM/TPU blends compatibilized with (a) MDI, (b) EPDM-*g*-MAH, (c) POE-*g*-MAH and (d) Joncryl. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

the interfacial interactions between POM and TPU. In Joncryl compatibilized POM/TPU blends, TPU particles exihibied smaller average size and narrower size distribution. And much enhanced interfacial interactions between POM and TPU were vividly demonstrated by impact force profile.

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